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Agricultural Fire Impacts on the Air Quality of Shanghai during Summer Harvesttime

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ABSTRACT

Agricultural fire is an important source of atmospheric carbonaceous aerosols. To better understand biomass burning emission originating from fire over Asian agriculture areas and its transport into the downwind atmosphere, aerosols and major trace gases were measured continuously from 22 May to 30 June at Shanghai during the summer harvesttime of 2009. Water-soluble K material contained in aerosols showed a clear day-to-day pattern with an average of 1.25 ± 1.48 μg/m^3. K ion loading and ratio of K ion to PM_{10} drastically increased during 'K event' days, accompanying with high PM_{10}, SO_2, and NO_2 levels. MODIS remote sensing fire map revealed about 80% agricultural fires occurred in the agriculture areas of Anhui, Jiangsu, Shandong and Henan provinces. Four potential source areas of agricultural fires, identified as Shanghai, Zhejiang, Jiangsu and Anhui provinces, had significant contributions to worsen the air quality of Shanghai during the harvest season.

Keywords: Agricultural fire; Air quality; Potential source.

INTRODUCTION

Smoke aerosols contain a large amount of partially oxidized organic material and black carbon (BC) or soot, which may have significant climate implications (IPCC, 2007). Second only to CO_2, in terms of direct climate forcing, BC is believed to be a principal component of global warming through absorption of solar radiation since its warming effect has been found to balance the net cooling effect of other anthropogenic aerosol constituents (Jacobson, 2001). Smoke aerosols originating from urban and industrial pollutions or some from wildfires have been shown to affect cloud microphysics, cloud reflection of sunlight to space, and the onset of precipitation (Andreae *et al.*, 2004). Delays in the onset of precipitation can increase cloud lifetime and thereby increase cloud cover (Resenfeld *et al.*, 2008). Most past researches on BC have been conducted in regions outside of Asia, but increasing recent studies within Asia show the importance of BC on that continent such as Asian Brown Cloud. Streets *et al.* (2001) estimated that BC emission in China (1342 Gg in 1995 and 1224 Gg in 2020), represents 30% of global anthropogenic emissions. Bond *et al.* (2004) have produced similar findings. Ohara *et al.* (2007) developed a new emission inventory for Asia for the period 1980–2020, and showed a rapid growth by 28% for BC and 30% for OC. The region is considered one of largest sources of smoke emission in the world.

Agricultural fire emits abundant gases and particles into the atmosphere, which causes serious local and regional air pollution during harvest seasons. In China, the field combustion of agricultural residues is a common way to eliminate waste after harvesting and a significant type of biomass burning. Measurements on gaseous and particulate emission factors from open burning of agricultural residues have been reported by Li *et al.* (2007) and Zhang *et al.* (2008). Some studies estimated that 17–25.6% of the total agricultural residue production, or 110–157.5 Tg of crop wastes were burned in the field in China every year (Cao *et al.*, 2006; Yan *et al.*, 2006). Cheng *et al.* (2006) carried out a continuous measurement of BC concentration from June 2004 to May 2005 at Hok Tsui, a site located downward of east Asia and China, and investigated the influence of southward outflow of continental pollution. Cao *et al.* (2003) profiled the characteristics of carbonaceous aerosols in the Pearl River Delta of China, and obtained the average

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concentrations of OC 14.7 μg/m^3 and EC 6.1 μg/m^3 in $PM_{2.5}$ during winter 2001. Cao *et al.* (2005) also characterized carbonaceous aerosols and apportioned the sources of EC and OC during the fall and winter of 2003 in Xi'an, China. Other measurements on carbonaceous aerosols and their distributions have been conducted in sites of Waliguan (Tang *et al.*, 1999), Beijing (Lou *et al.*, 2005), Tongliao (Li *et al.*, 2006) and Wusumu (Han *et al.*, 2008), etc. Li *et al.* (2009) used the dataset of agricultural fire sites derived from satellite remote sensing to analyze the impacts of field straw burning on urban air quality during summer wheat-harvested period.

Past studies have sufficiently portrayed the characteristics and role of anthropogenic aerosols in the atmosphere of Shanghai (Wang *et al.*, 2006), however, to our knowledge few has focused on the influence of carbonaceous aerosols from widespread rural biomass burning in Chinese agricultural areas. The purpose of this paper is to report agricultural fire impacts on the air quality of Shanghai using satellite remote sensing fire site data, and to determine the potential source contribution through tracing major outflow pathways.

MEASUREMENT

The atmospheric research station (31.3°N, 121.5°E) was established in 2005, and instruments were placed on the roof of one build about 20 m height above the ground in the campus of Fudan University in Shanghai of China.

A model ADI 2080 online analyzer of Monitoring of AeRosols and GAses (MARGA, Applikon Analytical B. V. Corp., Netherlands) with a PM_{10} inlet was used to measure the mass concentrations of major water-soluble aerosol inorganic ions and trace gases at the time resolution of 1 hour from 22 May to 30 June 2009. MARGA consists of a sampling box and an analytical box, with a particle collection efficiency of 99.7%. Ambient air is absorbed into the sampling box (flow 1 m^3/h) through the inlet, gaseous components are solubilized in the liquid film (0.0035% H_2O_2) formed by one Wet Rotating Denuder (WRD), and then particles in residual airflow go through the supersaturated steam (0.0035% H_2O_2, 120–140°C) erupted out from one Steam Jet Aerosol collector (SJAC) to be pooled into its collector. These two liquid samples flow through glass filters respectively, and are stored in syringe pumps of the analytical box ready for analyzing by Ion Chromatography (IC). MARGA has the capability of measuring the hourly average concentrations of NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+}, SO_4^{2-}, NO_3^-, Cl^-, HCl, HNO_2, SO_2, HNO_3, NH_3 in the atmosphere. During the field campaign, MARGA was calibrated using internal standard solution (LiBr) every week to verify accurate detecting limits and to ensure data quality. The daily average concentration of K ion was used here to identify field fire events as a proper indicator of biomass burning emission (Chow, 1995).

The daily fire site data during harvest seasons over Chinese agricultural areas between 19 May and 30 June 2009 were derived from the Ministry of Environmental Protection of China (Table 1), which was screened using ground land-use information on the basis of MODIS daily 1-km level 3 hotspot/fire products. Meteorological factors in Shanghai, including temperature, relative humidity (RH), wind speed and visibility, were downloaded from internet website http://www.wunderground.com. Daily average concentrations of SO_2, NO_2 and PM_{10} were from the Shanghai Environmental Protection Bureau (http://www.envir.gov.cn/airnews). To identify the influence of agricultural fire emissions from different regions on air quality at the sampling site, air mass trajectories were calculated using the HYSPLIT 4 model of the Air Resources Laboratory of NOAA (http://www.arl.noaa.gov/ready/hysplit4.html).

RESULTS AND DISCUSSION

Variation of Air Quality

It was known that water-soluble K ion is a proper indicator to be used in estimating biomass burning emission and tracing carbonaceous aerosol long-range transport in the atmosphere (Chow, 1995). The daily means of K ion concentrations for this study were evaluated from hourly average K^+ data collected by MARGA with PM_{10} sampler. Measured from 22 May to 30 June (Fig. 1), average K^+ concentration at Shanghai was 1.25 ± 1.48 μg/m^3. This is comparable to previous studies conduced at Beijing suburb by Zheng *et al.* (2005), with a yearly mean of 1.21 ± 0.97 μg/m^3, and at Beijing by Pu and Zhao (2006), with a autumnal mean of 1.49 μg/m^3. Average ratio

Table 1. Agricultural fire site number from 19 May to 30 June 2009.

Date	Fire number	Date	Fire number	Date	Fire number	Date	Fire number
19 May	4	30 May	217	10 June	226	21 June	31
20 May	39	31 May	41	11 June	16	22 June	128
21 May	64	1 June	231	12 June	372	23 June	11
22 May	0	2 June	36	13 June	434	24 June	32
23 May	109	3 June	364	14 June	274	25 June	2
24 May	1	4 June	299	15 June	153	26 June	9
25 May	83	5 June	561	16 June	13	27 June	11
26 May	2	6 June	56	17 June	37	28 June	3
27 May	268	7 June	28	18 June	2	29 June	17
28 May	56	8 June	3	19 June	35	30 June	4
29 May	175	9 June	4	20 June	4		

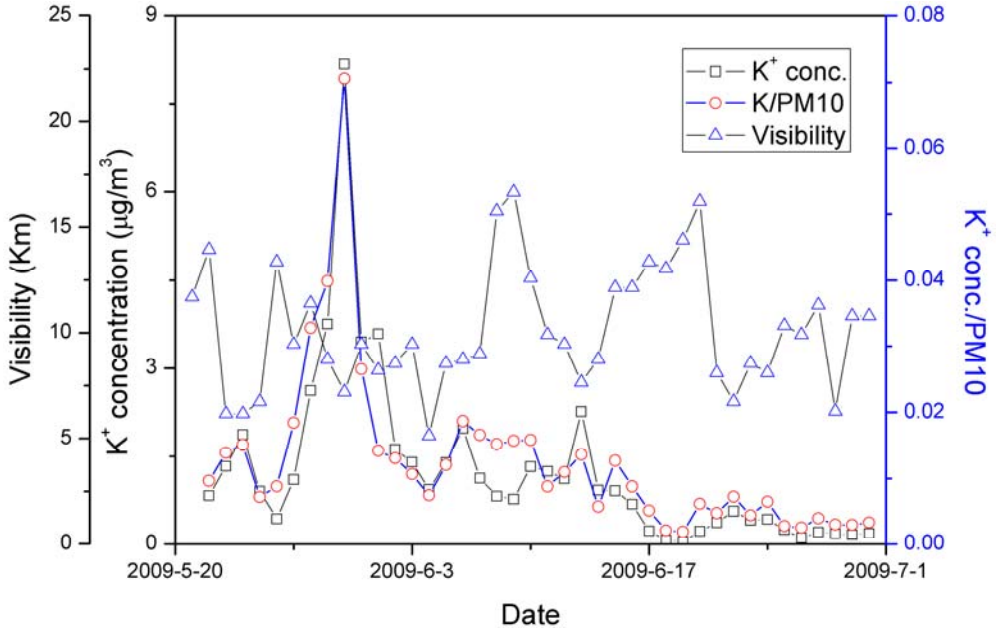

Fig. 1. Time series of daily average K^+ concentration, ratio of K^+ mass concentration to PM_{10}, and visibility in Shanghai.

of K^+ concentration to PM_{10} was 0.0125, less than K/TSP 0.0728 measured at Beijing in the autumn of 2004 (Pu and Zhao, 2006). A day-to-day variation of K^+ concentration was obvious, with the highest daily average on 30 May (8.19 μg/m^3) and the lowest value on 19 June (0.043 μg/m^3) (Fig. 1).

In order to profile the impacts of biomass burning emission from agricultural fire on air quality, we first scanned the daily K^+ loading data to pick up the 'K event', which was defined as periods when daily average K^+ concentration exceeded background value. Based on three criteria of K^+ concentration lower than average (1.25 μg/m^3), no agricultural fire occurrence in Shanghai suburb and visibility exceeding 10 km, the days disturbed by low carbonaceous aerosol loading were sieved out for clear periods. The average K^+ concentration of these selected clear days, in a magnitude of 0.3 μg/m^3, was served as the background against the 'K event', indicating contributions of other sources such as industrial and vehicle emissions in Shanghai, and remote biomass burning in domestic cooking and heating from upwind. As seen in Fig. 1, K ion loading and ratio K^+/PM_{10} drastically increased during event days. These criteria resulted in a somewhat subjective selection of events from the available records, but we expect that this will make the 'K event' more representative. Since there are no stable sources continuously emitting carbonaceous aerosols near the sampling location, except for traffic emissions, those events should reflect the local and long-range transport pollution from biomass burning related to agricultural fires.

Table 2 shows mean PM_{10}, chemical and meteorological conditions in various K^+ mass concentrations. High averages of PM_{10}, K^+, SO_2, NO_2 concentrations and K^+/PM_{10} appeared during the events compared to the background. For example, the average K^+ concentration (3.96 μg/m^3) of severe event days within 2.0–8.5 μg/m^3 was more than 24 times that of noevent days in 0.0–0.3 μg/m^3 range. In contrast, meteorological parameters of temperature, RH, wind speed and visibility almost reduced with K loading promoting. Wind speeds were usually low, with an average of 2.61 m/s for all event days, compared to the average 3.24 for noevent days. Meanwhile, visibility generally showed lower values during events than noevents. All of these are evidence of poor dispersion condition during carbonaceous aerosol events.

Agricultural Fire Event

To investigate outflows of biomass burning emission related to agricultural fire, the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1998) was used to trace 5-day back trajectories using K^+ as a marker for carbonaceous aerosols transported from agricultural regions. Fig. 2 shows 5-day back trajectories arriving at 150 m above the Shanghai sampling site for each day in high K^+ loading exceeding the background level during the summer harvest season of 2009. It was notable that the agricultural areas covered by the 5-day back trajectories arriving at the sampling site during daytime were similar with those during nighttime. As Fig. 2 shown, air parcel outflows arriving at the sampling site, prevailed in northerly and southerly pathways, had traveled a long distance over the Asian continent, and then toward coastal areas in eastern China, or across Fujian, Zhejiang provinces and even the Yangtze River Delta, and then southward to Shanghai. Streets *et al.* (2001) reported that BC emissions in China were concentrated in a west-to-east region across the agricultural heartland from Sichuan to Hebei provinces. In comparison with the BC emission inventory developed for China by Streets *et al.* (2001; 2003), most air masses arriving at Shanghai did not travel over those major BC emission source areas.

Table 2. Summary of chemical and meteorological conditions in various K^+ mass concentrations.

K^+ conc. (μg/m^3)	PM_{10} (mg/m^3)	SO_2 (mg/m^3)	NO_2 (mg/m^3)	Temp. (°C)	RH (%)	Wind speed (m/s)	Visibility (km)	K/PM_{10}
0.0–0.3	0.049	0.022	0.038	27.38	63.55	3.24	11.65	0.003
0.3–1.0	0.086	0.031	0.050	25.00	65.88	2.91	10.47	0.009
1.0–2.0	0.105	0.040	0.061	23.56	61.32	2.48	8.91	0.014
2.0–8.5	0.140	0.048	0.070	23.80	47.17	2.44	8.78	0.033
total	0.082	0.032	0.050	25.55	60.95	2.93	10.55	0.012

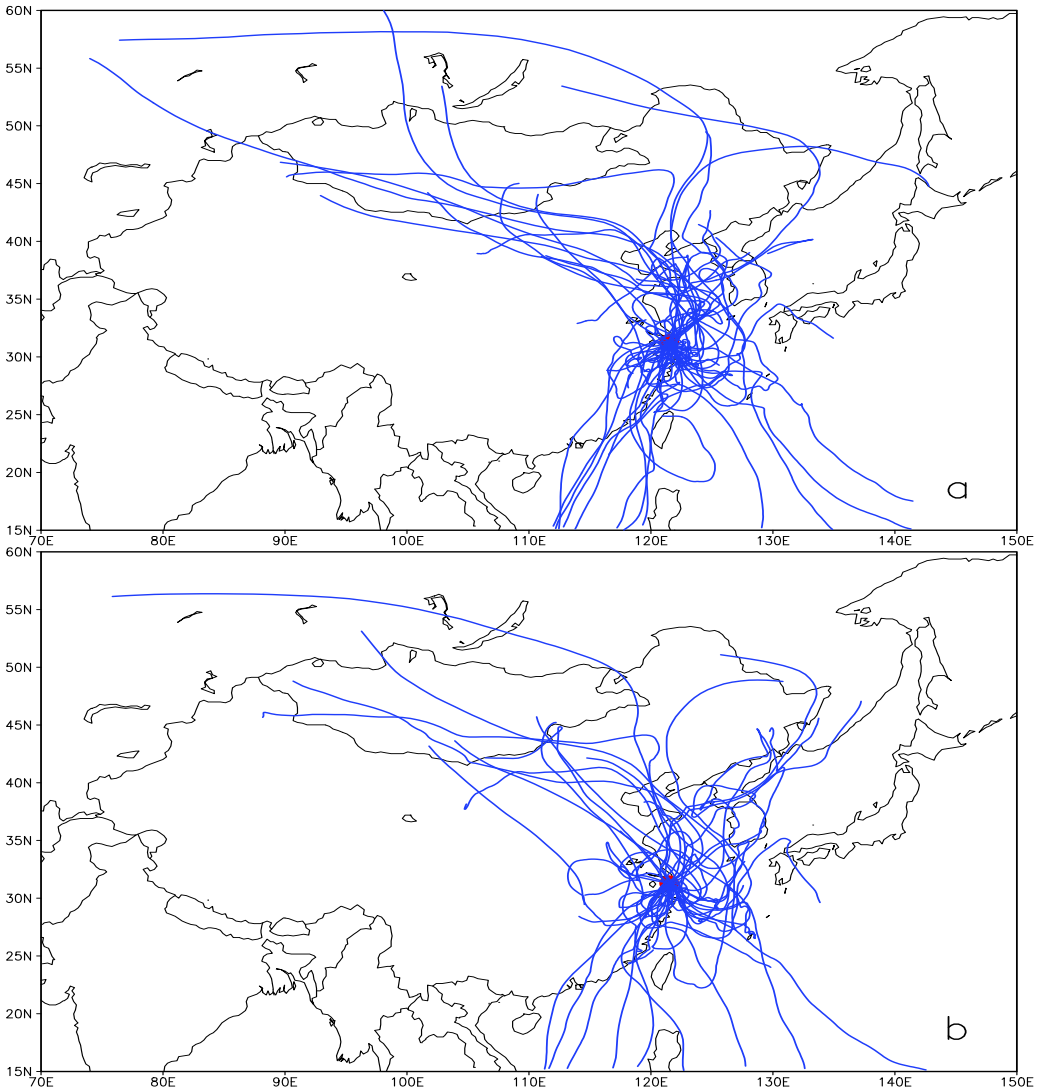

Fig. 2. Five-day air mass back trajectories at local time (a) 00:00 and (b) 12:00 during event days from 19 May to 30 June 2009.

MODIS of NASA EOS is an efficient sensor in space to provide information of fire distributions at 1 km resolution on the global scale. Fig. 3 gives a map of fire distributions derived from MODIS remote sensing products over primary Chinese agricultural areas from 19 May and 30 June 2009. These daily-summed fire sites scattered in regions partially corresponding with areas covered by the calculated air outflows during the summer harvest season (Fig. 2). Fig. 4 shows a statistics for all fire-site records collected in the whole harvest season. As can be seen, about 80% of agricultural fires occurred in Anhui, Jiangsu, Shandong and Henan provinces, and only less than 10% fires were found in Zhejiang, Shanghai, Hebei and other provinces, respectively.

Potential Source Contribution

In order to evaluate the potential source contribution of agricultural fires to air pollution in the atmosphere of Shanghai, correlation coefficient values were calculated between daily-summed fire numbers and daily-averaged K^+

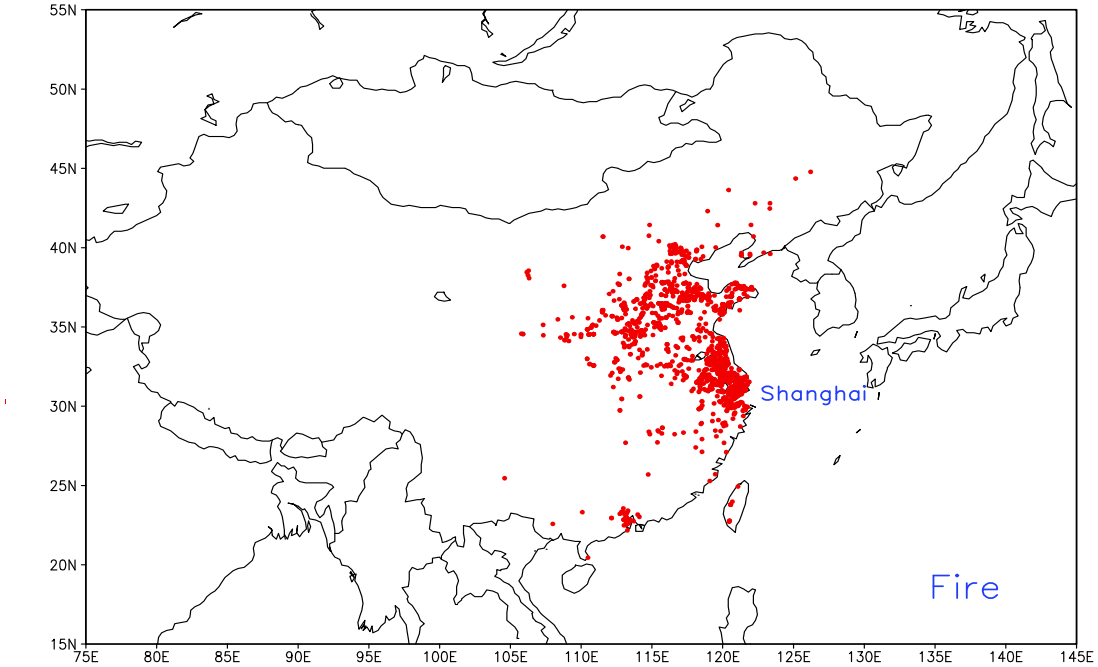

Fig. 3. Map of agricultural fire sites derived from satellite cloud-screened remote sensing from 19 May to 30 June 2009, corresponding to outflow covers in Fig. 2.

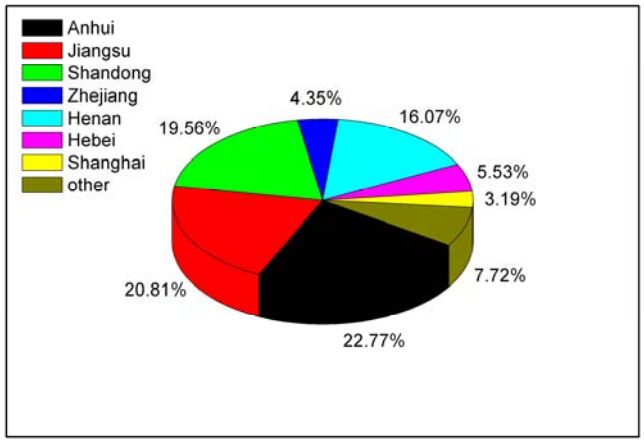

Fig. 4. Percentages of agricultural fires in regions of Anhui, Jiangsu, Shandong, Zhejiang, Henan, Hebei, and Shanghai.

mass concentrations. Moreover, back-to-fire interval, a period that biomass burning emission transported long range from source areas arriving over the sampling site along air masse outflows, was considered in these correlativity analyses. According to the results of relationship analysis (Table 3), four potential source areas of agricultural fires were identified as having important contributions to air quality at the sampling site. Shanghai, Zhejiang, Jiangsu and Anhui provinces constituted the four important source areas.

The biomass burning emission from agricultural fires, travelling for a period following air mass outflows, arrived at the sampling site to affect carbonaceous aerosol loading. As seen in table 3, for all event days with K^+ concentrations exceeding the background 0.3 μg/m^3, only the fire site records of Shanghai and Zhejiang provinces exhibited a high relativity with the K ion loadings in intraday and 1-day intervals. The fire site records of Jiangsu province were high correlated to the K ion loadings exceeding 1.0 μg/m^3 in 3-day interval, and the K ion loadings exceeding 3.0 μg/m^3 in intraday and 1-day intervals. Similar results were found in the fire site records of Anhui, Shandong, Guangdong, and Jiangxi provinces. All these indicated that agricultural fires which occurred in Shanghai suburb and Zhejiang province were the most important sources to rapidly and easily impact the air quality of Shanghai. Only during moderate and heavy events, agricultural fires which occurred in Jiangsu, Anhui, Shandong, Henan, and Guangdong provinces would force biomass burning emission to transport a long-range distance into the atmosphere of Shanghai. The potential source contribution of agricultural fires to Shanghai air quality is primary Shanghai and Zhejiang provinces, secondary Jiangsu and Anhui provinces, and final other provinces.

CONCLUSIONS

Measurements of major water-soluble inorganic ions contained in aerosols and trace gases were conducted at Shanghai from 22 May to 30 June during the summer harvesttime of 2009. The mean K^+ mass concentration during the whole campaign was 1.25 ± 1.48 μg/m^3. Throughout the sampling period, the minimum daily mean K ion loading was 0.043 μg/m^3 on 19 June, and the highest value occurred on 30 May (8.19 μg/m^3). Additional, outflow back trajectories clearly revealed that carbonaceous aerosols from biomass burning emission related to agricultural fires had significant impacts on the air quality of Shanghai.

Table 3. Correlation coefficients between daily average K^+ mass concentration and 1-day fire site number remote-sensed by satellite. The back-to-fire interval is the days from PM_{10} observation back to fire occurrence.

K^+ conc. ($\mu g/m^3$)	Back-to-fire interval	Shanghai	Zhejiang	Jiangsu	Anhui	Henan	Shandong	Guangdong-Jiangxi
0.3–8.5	0 day	**0.60**	**0.55**	0.02	–0.09	–0.15	0.04	–0.02
	1 day	**0.79**	**0.82**	0.18	–0.03	–0.13	–0.16	–0.03
	2 days	0.17	0.26	0.14	0.01	–0.13	–0.10	–0.12
1.0–8.5	0 day	**0.60**	**0.57**	–0.02	–0.25	–0.33	–0.05	–0.11
	1 day	**0.82**	**0.91**	0.34	–0.12	–0.24	–0.19	–0.14
	2 days	0.04	0.21	0.32	–0.07	–0.19	–0.22	–0.17
	3 days	–0.13	0.15	**0.75**	–0.06	–0.13	0.24	0.18
2.0–8.5	0 day	**0.55**	**0.54**	–0.08	–0.03	–0.35	–0.12	–0.30
	1 day	**0.84**	**0.90**	0.21	–0.26	–0.45	–0.20	–0.23
	2 days	–0.13	0.01	0.23	**0.75**	–0.44	–0.05	0.24
	3 days	–0.24	0.03	**0.77**	–0.07	–0.41	0.11	**0.86**
3.0–8.5	0 day	**0.50**	**0.50**	**0.65**	**0.63**	**0.87**	–0.09	–0.49
	1day	**0.84**	**0.89**	**0.83**	–0.05	–0.49	–0.16	–0.47
	2 days	–0.19	–0.06	0.20	**0.85**	–0.55	–0.11	0.22
	3 days	–0.30	–0.02	**0.81**	0.14	–0.33	**0.89**	**0.89**

Using back trajectories for the days with high K^+ concentrations, four potential source areas of agricultural fires of Shanghai suburb, Zhejiang, Jiangsu and Anhui provinces were identified as having important contributions to air quality at the sampling site. Most air masses arriving at Shanghai did not travel over the agricultural heartland in China. The biomass burning emission of agricultural fires in source areas of Shanghai suburb and Zhejiang provinces had more influence on K material in aerosols than source areas of Jiangsu, Anhui and other provinces.

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